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L8: Entry 1 of 4

File: PGPB

Mar 27, 2003

PGPUB-DOCUMENT-NUMBER: 20030060659

PGPUB-FILING-TYPE: new

DOCUMENT-IDENTIFIER: US 20030060659 A1

TITLE: Process for producing acrylic acid

PUBLICATION-DATE: March 27, 2003

## INVENTOR-INFORMATION:

NAME	CITY	STATE	COUNTRY	RULE-47
Yunoki, Hiromi	Himeji-shi		JP	

US-CL-CURRENT: 562/532

<a href="#">Full</a>	<a href="#">Title</a>	<a href="#">Citation</a>	<a href="#">Front</a>	<a href="#">Review</a>	<a href="#">Classification</a>	<a href="#">Date</a>	<a href="#">Reference</a>	<a href="#">Sequences</a>	<a href="#">Attachments</a>	<a href="#">Claims</a>	<a href="#">KMC</a>	<a href="#">Draw Desc</a>	<a href="#">Image</a>
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☐ 2. Document ID: US 20010021789 A1

L8: Entry 2 of 4

File: PGPB

Sep 13, 2001

PGPUB-DOCUMENT-NUMBER: 20010021789

PGPUB-FILING-TYPE: new

DOCUMENT-IDENTIFIER: US 20010021789 A1

TITLE: Process for producing acrolein and acrylic acid

PUBLICATION-DATE: September 13, 2001

## INVENTOR-INFORMATION:

NAME	CITY	STATE	COUNTRY	RULE-47
Tanimoto, Michio	Himeji-shi		JP	

US-CL-CURRENT: 562/534; 562/535

<a href="#">Full</a>	<a href="#">Title</a>	<a href="#">Citation</a>	<a href="#">Front</a>	<a href="#">Review</a>	<a href="#">Classification</a>	<a href="#">Date</a>	<a href="#">Reference</a>	<a href="#">Sequences</a>	<a href="#">Attachments</a>	<a href="#">Claims</a>	<a href="#">KMC</a>	<a href="#">Draw Desc</a>	<a href="#">Image</a>
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☐ 3. Document ID: US 6563000 B1

L8: Entry 3 of 4

File: USPT

May 13, 2003

US-PAT-NO: 6563000

DOCUMENT-IDENTIFIER: US 6563000 B1

TITLE: Process for producing acrylic acid

Full	Title	Citation	Front	Review	Classification	Date	Reference	Sequences	Attachments	Claims	KWIC	Draw Desc	Image
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☐ 4. Document ID: US 6028220 A

L8: Entry 4 of 4

File: USPT

Feb 22, 2000

US-PAT-NO: 6028220

DOCUMENT-IDENTIFIER: US 6028220 A

TITLE: Producing acrolein and acrylic acid using a supported dual activity molybdenum, iron, and bismuth based catalyst in a fixed bed multitubular reactor

Full	Title	Citation	Front	Review	Classification	Date	Reference	Sequences	Attachments	Claims	KWIC	Draw Desc	Image
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an inert carrier by a generally method described bellow and then calcined.

Brief Summary Text (43):

(In this formula, Mo, Bi, Ni, Co and Fe represent molybdenum, bismuth, nickel, cobalt and iron, respectively, Y is at least one element selected from the group of tin, zinc, tungsten, chromium, manganese, magnesium, antimony and titanium, Z is at least one element selected from the group of potassium, rubidium, thallium and cesium, and a, b, c, d, f, g, h and x represent the number of atoms of molybdenum, bismuth, nickel, cobalt, iron, Y, Z and oxygen, a=12, b=0.1 to 7, c+d=0.5 to 20, f=0.5 to 8, g=0 to 2, h=0 to 1 and x is determined by the oxidized condition of each element.) In the above definition, it is desirable that a=12, b=0.5 to 4, c+d=1 to 12, f=0.5 to 5, g=0 to 1 and h=0.01 to 0.5. The powder containing catalytically active component to be supported on a carrier, is prepared by a coprecipitation, spray drying or the like means, using nitrate, ammonium salt, hydroxide, oxide, acetate and the like salts of respective metal elements as the starting materials with no particular limitation. The powder containing catalytically active components is usually subjected to preliminary calcination at a temperature of from 200 to 600.degree.C., preferable 300 to 500.degree. C. for from 2 to 24 hours prior to its supporting on a carrier. The preliminary calcination is carried out preferably in the atmosphere of air or in a stream of nitrogen. The thus obtained powder by the preliminary calcination is called preliminarily calcined powder hereinafter. In addition, in production of a plurality of catalysts having different activities, when the catalysts having different activities are obtained by differing the catalytically active component itself or its composition ratio, it is also necessary that the composition of the catalytically active component of the supported catalysts (the case after preliminarily calcination and before supporting, the composition of the preliminarily calcined powder) (the composition of aforementioned formula) is differed, but the same composition of the catalytically active component is favorable because of the production becomes convenient.

Brief Summary Text (44):

When the preliminarily calcined powder described above is supported on a carrier, it is desirable to mix it with a molding additive and/or a strength improving agent. Illustrative examples of the molding additive include crystalline cellulose, starch, stearic acid and the like, and those of the strength improving agent include ceramic fibers, carbon fibers, whiskers and the like. The molding additive or strength improving agent is used in an amount of 30% by weight or less based on the preliminarily calcined powder. The molding additive and/or strength improving agent may be mixed in advance with the aforementioned preliminarily calcined powder prior to molding or, as will be described later, may be added to a molding machine simultaneously with or before or after the addition of the preliminarily calcined powder and the like.

Brief Summary Text (45):

It is desirable to use a binder when the preliminarily calcined powder is supported on the carrier. Illustrative examples of the binder include water, an alcohol, a polyhydric alcohol such as glycerol or the like or a mixture thereof. The binder is used in an amount of from 10 to 60% by weight based on the preliminarily calcined powder.

Brief Summary Text (47):

The powder that contains the catalytically active components (the powder containing preliminarily calcined powder, if necessary further containing a molding additive and/or a strength improving agent, to be referred to as catalytically active component-containing powder hereinafter) can be supported on the carrier by any method such as a tumbling granulation method, a method using a centrifugal fluidized bed coating apparatus and a wash coating method, with no particular limitation, but the tumbling granulation method is desirable when catalyst production efficiency and the like are taken into consideration. Illustratively, this is a method in which a fixed cylindrical container whose bottom part is equipped with a disc having even or irregular surface is used, and a carrier charged in the container is vigorously mixed through the repetition of rotation and revolution effected by high speed spinning of the disc, to which the catalytically active component-containing powder and, if necessary, a binder are added so that said powder is supported on the carrier.

Brief Summary Text (48):

A plurality of catalysts having different activities whose composition of the catalytically active component (the composition represented by aforementioned formula) is the same, may be obtained by differing a kind or amounts of a molding additive, a strength improving agent, and a binder which are added when the catalytically active component-containing powder is supported on the carrier. For example, when the ceramic fibers which are not burned out in the calcination after supporting, are used by differing their amount largely, concentration of the catalytically active component which are supported on the carrier is differed and thus catalysts having different activities may be obtained.

Brief Summary Text (49):

The ratio of the catalytically active component to the whole of the calcined catalyst may be differed with much extent of 5 to 80% by weight and the ratio is preferable 10 to 60% by weight and more preferable 20 to 55% by weight. When an any other component except of the catalytically active component to be supported on the carrier, for example, a molding additive or inert diluting material which are remained after the calcination, the ratio of the whole supported material (the ratio of the whole supported amount) is preferable within abovementioned ratio.

Brief Summary Text (51):

In this connection, the supported amount is represented by "weight of catalytically active components/(weight of catalytically active components+weight of carrier+weight of strength improving agent after calcination (optional component))", and is referred to as catalyst supported ratio hereinafter. The term "weight of catalytically active components" as used herein means weight of the catalytically active components after calcination and because of it being not so different to weight of the preliminarily calcined powder, the weight of the preliminarily calcined powder was used for calculation in this specification.

Brief Summary Text (52):

The calcination temperature after supporting catalytically active component-containing powder on the carrier, is preferably from 450 to 650.degree. C., more preferably from 480 to 600.degree. C.

Brief Summary Text (53):

Because the activity of the catalysts used in present invention becomes higher, when the calcination temperature or the calcination time is reduced, and becomes lower when calcination temperature or calcination time is increased, a plurality of catalysts having different activities are prepared by differing the calcination temperature and/or calcination time. This method is remarkably preferable for production of catalysts because a plurality of catalysts having different activities is obtained by same process before calcination.

Brief Summary Text (54):

The calcination time is generally from 3 to 30 hours, preferably from 4 to 15 hours. The preferred combination of calcination temperature and time is varied depending on the supported amount or the kind of catalytically active components and so on. For example, when the catalyst layer is divided into two parts a catalyst prepared by calcination at 500-650.degree. C. for 4-15 hours is preferably combined with a catalyst prepared by calcination at 450-550.degree. C. for 4-10 hours while the catalyst layer is divided into three parts a catalyst prepared by calcination at 500-650.degree. C. for 4-15 hours is preferably combined with a catalyst prepared by calcination at 500-580.degree. C. for 4-15 hours and a catalyst prepared by calcination at 450-550.degree. C. for 4-10 hours.

Brief Summary Text (55):

The difference of the calcination temperature between these catalysts, is generally less than 100.degree. C., preferably 60.degree. C. In case that the activity is adjusted by differing the calcination time, the calcination temperature may be same, but it is preferable to combine a plurality of catalysts having different activities obtained by calcining at difference of the calcination temperature being above 5.degree. C., preferably 10.degree. C., more preferably 15.degree. C.

Brief Summary Text (58):

Inner diameter of the reaction tube is generally from about 15 to 50 mm. For example, when the inner diameter of reaction tube is 21 to 27 mm in that case, it is desirable to use a combination of catalysts having a particle size of 3.5 to 8 mm whose activities are controlled by adjusting the supported amount in the catalysts to 15 to 60% by weight, and the calcination temperature to 480 to 580.degree. C.

Brief Summary Text (59):

The production method of the present invention may be applied to a once-through (one-pass) operation method or a recycling method, and can be carried out under conditions generally used on them. For example, the reaction to obtain acrolein and acrylic acid may be carried out by introducing a mixture gas consisting of 1 to 10% by volume, preferably 4 to 9% by volume, of propylene as the starting material, 3 to 20% by volume, preferably 4 to 18% by volume, of molecular oxygen, 0 to 60% by volume, preferably 4 to 50% by volume, of water vapor and 20 to 80% by volume, preferably 30 to 60% by volume, of an inert gas (nitrogen, carbon dioxide or the like) onto the aforementioned catalysts at a temperature of 250 to 450.degree. C., under a pressure of atmospheric pressure to 10 atm and at a space velocity (=material gas flow volume/apparent volume of packed catalysts) of 300 to 5,000 hr.sup.-1.

Brief Summary Text (61):

When the reaction is carried out according to the present invention by supporting a catalytically active component-containing powder on a carrier, calcining the resultant and then packing a plurality of the thus obtained catalysts having different activities in each of a plurality of reaction zones arranged in the reaction tube axial direction in such order that the catalyst with lower activity is placed from the inlet of the starting material, the danger of causing runaway reaction due to the generation of hot spots or heat accumulation at the hot spot can be avoided and formation of by-products due to over oxidation reaction can be prevented, even under high load reaction conditions, so that the intended acrolein and acrylic acid can be obtained with high selectivity and high yield. The productivity can also be improved markedly, because deterioration of the catalyst due to locally exceeded thermal load can be prevented and the catalyst therefore can be used stably for a prolonged period of time.

Brief Summary Text (62):

Thus, the production method of the present invention is a markedly useful method for the production of acrolein and acrylic acid.

Detailed Description Text (5):

Total selectivity (mol %)=(mol number of formed acrolein and acrylic acid)/(mol number of reacted propylene).times.100

Detailed Description Text (6):

Yield (mol %)=(mol number of formed acrolein or acrylic acid)/(mol number of supplied propylene).times.100

Detailed Description Text (11):

The thus formed suspension was dried using a spray dryer and subjected to 3 hours of preliminary calcination at 440.degree. C., thereby obtaining 570 g of preliminarily calcined powder. Thereafter, 200 g of the preliminarily calcined powder was mixed with 10 g of crystalline cellulose as a molding additive, and thus a mixture of the preliminarily calcined powder and the crystalline cellulose was obtained.

Detailed Description Text (13):

The active component-supported particles were dried at room temperature for 15 hours and then calcined at 560.degree. C. for 5 hours in the flow of air to obtain a catalyst (1). The catalyst (1) was found to have an average particle size of 4.0 mm, and the composition of its catalytically active components, excluding oxygen, was Mo=12, Bi=1.7, Ni=2.8, Fe=1.8, Co=5.2 and K=0.1 in atomic ratio.

Detailed Description Text (15):

A 300 g portion of the preliminarily baked powder obtained in Preparation of catalyst-1 was mixed with 15 g of crystalline cellulose as a molding additive, and thus obtained a mixture of the preliminarily clined powder and the crystalline cellulose was obtained.

Detailed Description Text (17):

The thus obtained active component-supported particles were dried at room temperature for 15 hours and then calcined at 520.degree. C. for 5 hours in the flow of air to obtain a catalyst (2). The catalyst (2) was found to have an average particle size of 4.1 mm, and the composition of its catalytically active components, excluding oxygen, was Mo=12, Bi=1.7, Ni=2.8, Fe 1.8, Co=5.2 and K=0.1 in atomic ratio.

Detailed Description Text (19):

The aforementioned catalysts (1) and (2) were packed in a reaction tube of 21 mm in inner diameter and 5 m in length made of stainless steel (SUS 304) and equipped with a thermocouple, in respective packed layer lengths of 105 cm and 295 cm at central area of the reaction tube starting from the starting material gas inlet toward the outlet. While keeping the reaction bath temperature at 337.degree. C. and the catalyst layer inlet pressure at 1.53 kg/cm.sup.2 G, the reaction was carried out by passing a mixture gas consisting of 8% by volume of propylene 14% by volume of oxygen, 25% by volume of water vapor and 53% by volume of nitrogen through the tube at a space velocity of 1,860 hr<sup>-1</sup>. In this case, maximum temperatures of the catalyst layers starting from the inlet were 388.degree. C. and 400.degree. C., respectively, the propylene conversion ratio was 97.1%, the acrolein yield was 80.9%, the acrylic acid yield was 8.1% and the total selectivity for acrolein and acrylic acid was 91.7%, and decrease in reaction performance was not observed even after continuation of the reaction for 1,000 hours or more.

Detailed Description Text (22):

A catalyst (3) was obtained in the same manner as described in Preparation of catalyst-1, except that the calcining temperature was changed to 540.degree. C. The catalyst (3) was found to have an average particle size of 4.0 mm, and the composition of its catalytically active components, excluding oxygen, was Mo=12, Bi=1.7, Ni=2.8, Fe=1.8, Co=5.2 and K=0.1 in atomic ratio.

Detailed Description Text (24):

The aforementioned catalysts (3) and (2) were packed in a reaction tube of 21 mm in inner diameter and 5 m in length made of stainless steel (SUS 304) and equipped with a thermocouple, in respective packed layer lengths of 105 cm and 295 cm starting from the starting material gas inlet toward the outlet. While keeping the reaction bath temperature at 333.degree. C. and the catalyst layer inlet pressure at 1.17 kg/cm.sup.2 G, the reaction was carried out by passing a mixture gas consisting of 8% by volume of propylene, 14% by volume of oxygen, 25% by volume of water vapor and 53% by volume of nitrogen through the tube at a space velocity of 1,550 hr<sup>-1</sup>. In this case, maximum temperatures of the catalyst layers starting from the inlet were 401.degree. C. and 373.degree. C., respectively, the propylene conversion ratio was 98.1%, the acrolein yield was 81.3%, the acrylic acid yield was 8.6% and the total selectivity for acrolein and acrylic acid was 91.6%, and decrease in reaction performance was not observed even after continuation of the reaction for 1,000 hours or more.

Detailed Description Text (27):

A 100 g portion of the preliminarily calcined powder obtained in Preparation of catalyst-1 was mixed with 5 g of crystalline cellulose as a molding additive, and thus a mixture of the preliminarily calcined powder and the crystalline cellulose was obtained.

Detailed Description Text (29):

The thus obtained active component-supported particles were dried at room temperature for 15 hours and then calcined at 520.degree. C. for 5 hours in the flow of air to obtain a catalyst (4). The catalyst (4) was found to have an average particle size of 4.3 mm, and the composition of its catalytically active components, excluding oxygen, was Mo=12, Bi=1.7, Ni=2.8, Fe=1.8, Co=5.2 and K=0.1 in atomic ratio.

Detailed Description Text (31):

A 150 g portion of the preliminarily calcined powder obtained in Preparation of catalyst-1 was mixed with 7.5 g of crystalline cellulose as a molding additive, and thus a mixture of the preliminarily calcined powder and the crystalline cellulose was obtained.

art, thereby providing a method by which acrolein and acrylic acid can be produced from propylene with a high efficiency.

Brief Summary Text (16):

That is, it contemplates providing a method in which acrolein and acrylic acid are produced by subjecting propylene to vapor phase catalytic oxidation under high load reaction conditions, which is a simple and easy method that can effect the production stably for a prolonged period of time by inhibiting generation of hot spots or excessive heat accumulation at the hot spot of the catalyst layer, obtaining the products of interest with high yields and preventing the catalyst from its deterioration by thermal load.

Brief Summary Text (18):

In the exothermic reaction such as the case of the vapor phase catalytic oxidation reaction, catalytically active components are generally used by molding them into various shapes which components are mostly occupied by the catalytically active components. Since the catalyst can be regarded as the reaction field of vapor phase catalytic oxidation, the exothermic reaction occurs exactly on the catalyst. Thus, the heat generated by the reaction is concentrated to induce generation of hot spots. In view of the above, the inventors of the present invention have conducted intensive studies with the aim of obtaining the products of interest stably for a prolonged period of time by avoiding the concentration of heat of reaction generated on the catalyst, and found as a result of the efforts that the aforementioned object can be achieved when a plurality of supported catalysts, having different activities, which are prepared by supporting a suitable amount of a powder that contains catalytically active components to be supported (coated) on an inert carrier and by differing the calcining procedure at preparing the catalysts or the supporting amount of the catalyst, are used by arranging them in a specified manner.

Brief Summary Text (20):

(1) a method for producing acrolein and acrylic acid by carrying out vapor phase catalytic oxidation of propylene with molecular oxygen or a gas containing molecular oxygen using a oxidation catalysts comprising Mo, Bi and Fe as an essentially element and a fixed bed multitubular reactor, which comprises

Brief Summary Text (24):

(2) the method according to the above item (1) wherein plural catalysts having different activities, are prepared whose activity is controlled by means of the calcining treatment in the catalysts preparing process,

Brief Summary Text (26):

(4) the method according to the above item (2) wherein the supported plural catalysts having different activities, are calcined in different temperature selected from 450.about.650.degree. C. for every supported catalyst,

Brief Summary Text (28):

(6) the method according to the above item (5) wherein the supported plural catalysts having different activities, are obtained by using a molding additive and/or a strength improving agent when the supported catalysts are prepared,

Brief Summary Text (36):

(12) the method according to any one of the above items (1), (2), (4), (6) and (8) wherein the plural supported catalysts having different activities, which are obtained by calcining the lower temperature or whose supporting amount are the more, are arranged at the nearer position of the outlet.

Brief Summary Text (41):

The oxidation catalyst component to be supported on the carrier is not limited whichever the components comprises molybdenum, bismuth and iron as generally being used for production of acrolein and acrylic acid. Such catalyst component are much disclosed in the U.S. Pat. Nos. 3,454,630, 3,778,386 and Japanese patents described before. The supported catalysts to be used in the present invention which are supporting the oxidation catalysts component disclosed in those prior arts, on an inert carrier can be obtained usually by a method in which catalytically active components that contain a powder of catalytically active components are supported on



Detailed Description Text (33):

The thus obtained active component-supported particles were dried at room temperature for 15 hours and then calcined at 520.degree. C. for 5 hours in the flow of air to obtain a catalyst (5). The catalyst (5) was found to have an average particle size of 4.5 mm, and the composition of its catalytically active components, excluding oxygen, was Mo=12, Bi=1.7, Ni=2.8, Fe=1.8, Co=5.2 and K=0.1 in atomic ratio.

Detailed Description Text (35):

A 200 g portion of the preliminarily calcined powder obtained in Preparation of catalyst-1 was mixed with 10 g of crystalline cellulose as a molding additive, and thus a mixture of the preliminarily calcined powder and crystalline cellulose was obtained.

Detailed Description Text (37):

The thus obtained active component-supported particles were dried at room temperature for 15 hours and then calcined at 520.degree. C. for 5 hours in the flow of air to obtain a catalyst (6). The catalyst (6) was found to have an average particle size of 4.5 mm, and the composition of its catalytically active components, excluding oxygen, was Mo=12, Bi=1.7, Ni=2.8, Fe=1.8, Co=5.2 and K=0.1 in atomic ratio.

Detailed Description Text (39):

The aforementioned catalysts (4), (5) and (6) were packed in a reaction tube of 27 mm in inner diameter and 5 m in length made of stainless steel (SUS 304) and equipped with a thermocouple, in respective packed layer lengths of 100 cm, 100 cm and 150 cm starting from the starting material gas inlet toward the outlet. While keeping the reaction bath temperature at 334.degree. C. and the catalyst layer inlet pressure at 1.35 kg/cm.sup.2 G, the reaction was carried out by passing a mixture gas consisting of 7% by volume of propylene, 13% by volume of oxygen, 10% by volume of water vapor and 70% by volume of nitrogen through the tube at a space velocity of 1,800 hr<sup>-1</sup>. In this case, maximum temperatures of the catalyst layers starting from the inlet were 404.degree. C., 385.degree. C. and 352.degree. C., respectively, the propylene conversion ratio was 96.6%, the acrolein yield was 84.2%, the acrylic acid yield was 6.2% and the total selectivity for acrolein and acrylic acid was 93.6%, and decrease in reaction performance was not observed even after continuation of the reaction for 1,000 hours or more.

Detailed Description Text (41):

The reaction was carried out in the same manner as described in Example 3, except that the space velocity was changed to 1,500 hr<sup>-1</sup>, the reaction bath temperature to 332.degree. C. and the catalyst layer inlet pressure to 1.1 kg/cm.sup.2 G. In this case, maximum temperatures of the catalyst layers were 398.degree. C., 381.degree. C. and 350.degree. C., respectively, the propylene conversion ratio was 96.4%, the acrolein yield was 84.3%, the acrylic acid yield was 6.0% and the total selectivity for acrolein and acrylic acid was 93.6%, and decrease in the reaction performance was not observed even after continuation of the reaction for 1,000 hours or more.

Detailed Description Text (44):

A 200 g portion of the preliminarily calcined powder obtained in Preparation of catalyst-1 was mixed with 10 g of crystalline cellulose as a molding additive, and thus a mixture of the preliminarily calcined powder and the crystalline cellulose was obtained.

Detailed Description Text (46):

The thus obtained active component-supported particles were dried at room temperature for 15 hours and then calcined at 550.degree. C. for 5 hours in the flow of air to obtain a catalyst (7). The catalyst (7) was found to have an average particle size of 4.5 mm, and the composition of its catalytically active components, excluding oxygen, was Mo=12, Bi=1.7, Ni=2.8, Fe=1.8, Co=5.2 and K=0.1 in atomic ratio.

Detailed Description Text (48):

The aforementioned catalysts (7) and (6) were packed in a reaction tube of 21 mm in inner diameter and 5 m in length made of stainless steel (SUS 304) and equipped with a thermocouple, in respective packed layer lengths of 105 cm and 295 cm starting from the starting material gas inlet toward the outlet. While keeping the reaction bath

temperature at 339.degree. C. and the catalyst layer inlet pressure at 1.81 kg/cm.sup.2 G, the reaction was carried out by passing a mixture gas consisting of 8% by volume of propylene, 14% by volume of oxygen, 25% by volume of water vapor and 53% by volume of nitrogen through the tube at a space velocity of 1,860 hr-1. In this case, maximum temperatures of the catalyst layers starting from the inlet were 406.degree. C. and 397.degree. C., respectively, the propylene conversion ratio was 97.4%, the acrolein yield was 79.9%, the acrylic acid yield was 8.9% and the total selectivity for acrolein and acrylic acid was 91.2%, and decrease in reaction performance was not observed even after continuation of the reaction for 1,000 hours or more.

Detailed Description Text (50):

According to the present invention, run away reaction caused by the generation of hot spot and over oxidation reaction can be avoided and constant operation can be made for a prolonged period of time without employing industrially disadvantageous methods, even under high load reaction conditions in which concentration of the starting material is increased and/or the space velocity is increased, so that the method of the present invention is an outstandingly usable method for production of acrolein and acrylic acid.

Issued US Original Classification (1):

562/546

Current US Original Classification (1):

562/546

Current US Cross Reference Classification (1):

562/531

Current US Cross Reference Classification (2):

562/532

Current US Cross Reference Classification (3):

562/545

Issued US Cross Reference Classification (1):

562/531

Issued US Cross Reference Classification (2):

562/532

Issued US Cross Reference Classification (3):

562/545

Field of Search Class/SubClass (1):

562/531

Field of Search Class/SubClass (2):

562/532

Field of Search Class/SubClass (3):

562/534

Field of Search Class/SubClass (4):

562/545

Field of Search Class/SubClass (5):

562/546

US Reference US Original Classification (1):

562/537

US Reference Group (1):

5276178 19940100 Onodera et al. 562/537

## CLAIMS:

1. A method for producing acrolein and acrylic acid by carrying out vapor phase catalytic oxidation of propylene with molecular oxygen or a gas containing molecular oxygen using oxidation catalysts comprising Mo, Bi and Fe as an element and a fixed bed multitubular reactor, which comprises
  - a) using a plurality of supported catalysts having different activities depending on an amount of catalyst supported on a carrier,
  - b) setting a plurality of catalyst layers within a reaction tube, which is formed by dividing it into plural portions in the tube axial direction, and
  - c) arranging the catalyst layers in such order that the activity of the catalyst in each said layer becomes high toward the outlet from the inlet of the material gas in the reaction tube axial direction.
4. The method according to claim 3 wherein the supported plural catalysts having different activities, are obtained by using a molding additive and/or a strength improving agent when the supported catalysts are prepared.
10. The method according to any one of claims 1 or 4 wherein of the plural supported catalysts having different activities, the catalyst which are obtained by calcining at a lower temperature or whose supporting amount are higher, are arranged nearer said outlet.
14. The method according to claim 6, wherein of the plural supported catalysts having different activities, the catalyst which are obtained by calcining at a lower temperature or whose supporting amount are higher are arranged nearer said outlet.

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L8: Entry 4 of 4

File: USPT

Feb 22, 2000

DOCUMENT-IDENTIFIER: US 6028220 A

TITLE: Producing acrolein and acrylic acid using a supported dual activity molybdenum, iron, and bismuth based catalyst in a fixed bed multitubular reactorAbstract Text (1):

The present invention concerns a method for producing acrolein and acrylic acid by carrying out vapor phase catalytic oxidation of propylene with molecular oxygen or a gas containing molecular oxygen using a oxidation catalysts comprising Mo, Bi and Fe as an essentially element and a fixed bed multitubular reactor, which comprises

Abstract Text (2):

a) using a plurality of supported catalysts having different activities, which was obtained, for example, by different calcination method in the production process of the supported catalysts,

Abstract Text (5):

According to the present invention, the generation of hot spot can be avoided and over oxidation reaction can be avoided and the acrolein and acrylic acid are produced constantly for a prolonged period of time.

Brief Summary Text (2):

This invention relates to a method in which acrolein and acrylic acid are produced by carrying out vapor phase catalytic oxidation of propylene with molecular oxygen or a gas containing molecular oxygen using a fixed bed multitubular reactor.

Brief Summary Text (4):

A number of complex oxide catalysts containing molybdenum, bismuth and iron have already been proposed for use in the production of acrolein and acrylic acid by a method in which propylene is subjected to vapor phase catalytic oxidation, and some of them are now industrially used. Their typical examples include those which are disclosed in Japanese Patent Publication (KOKOKU) No. 47-27490 (1972), Japanese Patent Publication (KOKOI(U) No. 47-42241 (1972) and Japanese Patent Publication (KOKOKU) No. 48-1645 (1973).

Brief Summary Text (5):

However, industrial production of acrolein or acrylic acid using these catalysts causes various problems.

Brief Summary Text (6):

One of these problems is generation of a local abnormally high temperature part (hot spot) in the catalyst layer. Generation of the hot spot is caused by the exothermic reaction of said vapor phase catalytic reaction. In order to improve productivity in the industrial production of acrolein and acrylic acid, a means is generally employed in which the concentration of the starting material propylene is increased or the space velocity of the reaction gas is increased, but heat accumulation at the hot spot is increased under such high load reaction conditions. Increase in heat accumulation at the hot spot causes shortened catalyst life, increased formation of by-products due to over oxidation reaction and, in the worst case, runaway reaction.

Brief Summary Text (15):

The present invention contemplates resolving the aforementioned problems in the prior